

Breakdown of Magnons in a Strongly Spin-Orbital Coupled Magnet

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The description of quantized collective excitations stands as a landmark in the quantum theory of condensed matter. A prominent example occurs in conventional magnets, which support bosonic magnons - quantized harmonic fluctuations of the ordered spins. In striking contrast is the recent discovery that strongly spin-orbital coupled magnets, such as α -RuCl₃, may display a broad excitation continuum inconsistent with conventional magnons. Unraveling the nature of this continuum, however, remains challenging due to incomplete knowledge of the underlying interactions. While the most discussed explanation refers to a coherent continuum of fractional excitations analogous to the celebrated Kitaev spin-liquid, we present here a more general scenario. We propose the observed continuum represents incoherent excitations originating from strong magnetic anharmonicity that naturally occurs in such materials. This scenario fully explains the observed inelastic magnetic response of α -RuCl₃ and reveals the presence of nontrivial excitations in such materials extending well beyond the Kitaev state.

Introduction – From magnons in ordered magnets to phonons in periodic crystals, the appearance of bosonic collective excitations is ubiquitous in condensed phases [1]. For this reason, special attention is given to those states supporting more exotic collective modes, for which the conventional paradigm breaks down. In the context of magnetic phases, the breakdown of magnons is commonly thought to require closeness to an unconventional state such as a quantum spin liquid [2–4]. A notable example occurs in Kitaev’s exactly solvable honeycomb model [5], for which strongly anisotropic and bond-dependent interactions fractionalize conventional spin excitations into Majorana spinons and fluxes. This Kitaev state has recently risen to prominence due to the suggestion that it may be realized in heavy metal $4d^5$ and $5d^5$ insulators via a specific interplay between the crystal field and strong spin-orbit coupling [6] and, consequently, a variety of candidate materials based on Ir⁴⁺ and Ru³⁺ have been intensively explored [7]. Encouragingly, evidence of a continuum of magnetic excitations inconsistent with conventional magnons was found in the majority of such materials, including the two-dimensional (2D) honeycomb Na₂IrO₃ [8, 9] and α -RuCl₃ [10–13], as well as the 3D analogues β -, γ -Li₂IrO₃ [14], despite possessing all of them magnetically ordered ground states.

While the observed excitation continua in the above systems has been interpreted in terms of signatures of the Kitaev state, the low-symmetry crystalline environment of the real materials also allows various additional interactions beyond Kitaev’s model [15–17], which are thought to be large based on both experimental [18, 19] and theoretical [17, 20, 21] considerations. In this sense, understanding the mechanism for the breakdown of magnons and appearance of a broad continuum of magnetic excitations remains a key challenge.

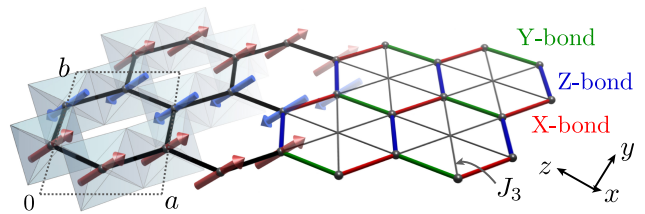


FIG. 1. From material to model within the honeycomb ab -layer of α -RuCl₃. Crystal axes are labelled with respect to the $C2/m$ structure. Illustrated are the RuCl₆ octahedra, magnetic zigzag ordering pattern, and definition of the underlying magnetic interactions.

In this work, we study as a representative case α -RuCl₃, which forms a layered 2D honeycomb lattice, and displays zigzag magnetic order below $T_N \sim 7$ K as shown in Fig. 1 [12, 13, 22]. We specifically address the recent inelastic neutron scattering (INS) measurements, which have revealed low-energy magnons [23] coexisting with an intense excitation continuum [12]. The latter continuum possesses a distinctive six-fold star shape in momentum space, and large intensity at the 2D Γ -point over a wide energy range $E = 2 - 15$ meV [12]. To resolve the nature of this continuum, we take two complementary approaches. We first theoretically investigate the neutron spectra over a range of relevant magnetic interactions in order to determine the correct spin Hamiltonian for α -RuCl₃, which has been an intense subject of recent discussion [17, 24–27]. Second, we identify the conditions that lead to the breakdown of conventional magnons in the presence of strongly anisotropic and frustrated interactions, revealing that nontrivial excitations naturally persist well beyond the Kitaev spin-liquid.

Results – Based on previous *ab-initio* studies [17, 24–

27], the largest terms in the spin Hamiltonian of α - RuCl_3 are generally expected to include nearest neighbour Heisenberg J_1 , Kitaev K_1 , and off-diagonal Γ_1 couplings, supplemented by a 3rd neighbour Heisenberg J_3 term:

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \sum_{\langle\langle i,j \rangle\rangle} J_3 \mathbf{S}_i \cdot \mathbf{S}_j \quad (1)$$

where the bond-dependent labels $\{\alpha, \beta, \gamma\}$ are, for example, equal to $\{x, y, z\}$ for the Z-bond shown in Fig. 1. The phase diagram of this model has been discussed previously [16, 17, 25, 28], and is further detailed in the supplemental material; here we review the key aspects.

In the limit where J_1, Γ_1 , and J_3 vanish, the ground state is a gapped \mathbb{Z}_2 spin-liquid for either positive or negative K_1 , as demonstrated in Kitaev's seminal work [5]. Small deviations from these limits induce various magnetically ordered states. The simplest perturbation is the introduction of a finite J_1 , which yields the well-studied (J_1, K_1) nearest neighbour Heisenberg-Kitaev (nnHK) model. This model hosts zigzag order consistent with α - RuCl_3 for $K_1 > 0, J_1 < 0$, as shown in Fig. 2(a) [29]. Accordingly, previous analysis of the powder INS experiments within the context of the nnHK model [13], suggested that $K_1 \sim +7$ meV, and $|J_1/K_1| \sim 0.3 - 0.7$ for α - RuCl_3 . On this basis, the broad excitation continua observed experimentally have been interpreted in terms of proximity to the $K_1 > 0$ Kitaev spin-liquid [12, 13]. However, the further introduction of $\Gamma_1, J_3 > 0$ interactions in Eq. (1) significantly expands the experimentally relevant region, as both interactions generally stabilize zigzag order (Fig. 3(a)). Indeed, recent *ab-initio* studies [17, 24–27] have suggested that the zigzag order emerges from $J_1 \sim 0, K_1 < 0, \Gamma_1 > 0$, and $J_3 > 0$, with $|\Gamma_1/K_1| \sim 0.5 - 1.0$, and $|J_3/K_1| \sim 0.1 - 0.5$, indicating significant deviations from both Kitaev's original model and the region identified by initial experimental analysis. Before discussing the origin of the excitation continua, it is therefore crucial to first pinpoint the relevant interaction parameters for α - RuCl_3 .

In order to address this issue directly, we have computed the neutron scattering intensity $\mathcal{I}(\mathbf{k}, \omega)$ for a variety of interactions within the zigzag ordered phase via both linear spin-wave theory (LSWT) and exact diagonalization (ED). For the latter case, we combine results from various periodic 20- and 24-site clusters compatible with the zigzag state in order to probe a wider range of \mathbf{k} -points, (see Methods section). Full results for the complete range of models are presented in the supplemental material; here we highlight the key aspects in the form of Fig. 2 and 3, which show detailed ω - and \mathbf{k} -dependence of $\mathcal{I}(\mathbf{k}, \omega)$ for two representative sets of interactions, along with the evolution of the spectra on moving towards the

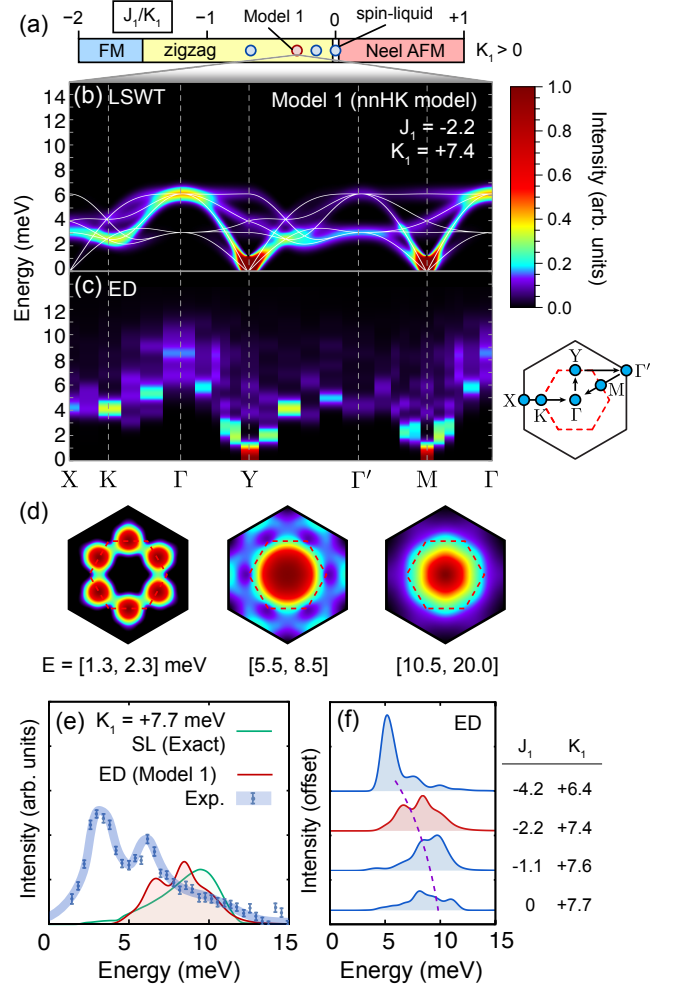


FIG. 2. Computed neutron scattering intensity $\mathcal{I}(\mathbf{k}, \omega)$ within the nnHK model. (a) Phase diagram for $K_1 > 0$ as a function of $|J_1/K_1|$, showing positions of interactions shown in (f). (b-d) Detailed results for Model 1: (b) $\mathcal{I}(\mathbf{k}, \omega)$ computed via LSWT; results are averaged over the three zigzag ordering wavevectors, parallel to the X, Y, and Z bonds. (c) ED results, combining data from several 20- and 24-site periodic clusters (see Methods). (d) ED \mathbf{k} -dependence of $\mathcal{I}(\mathbf{k}, \omega)$ integrated over the indicated energies, as obtained from a single 24-site cluster respecting all symmetries of Eq. (1) (see Methods). (e) Comparison of Γ -point intensities for the $K_1 > 0$ spin-liquid (exact results [30, 31]), Model 1 (ED), and the experimental data for α - RuCl_3 [12]. (f) Evolution of the ED Γ -point intensity with decreasing $|J_1/K_1|$, showing absence of low-energy intensity close to the $K_1 > 0$ spin-liquid. For all spectra, a Gaussian broadening of 0.5 meV has been applied.

$K_1 > 0$ or $K_1 < 0$ spin-liquid. Within the (J_1, K_1) nnHK model, we focus on Model 1 ($J_1 = -2.2, K_1 = +7.4$ meV; $|J_1/K_1| = 0.3$), which lies on the border of the initially identified region in Ref. [12], close to the spin liquid. Beyond the nnHK model, we consider Model 2 ($J_1 = -0.5, K_1 = -5.0, \Gamma_1 = +2.5, J_3 = +0.5$ meV) for which parameters have been guided by recent *ab-initio*

studies [17, 24–27], and further optimized to improve agreement with the experimental spectra.

We begin by analyzing the spectra $\mathcal{I}(\mathbf{k}, \omega)$ within the zigzag phase of the (J_1, K_1) nnHK model, starting with Model 1 (Fig. 2). Despite proximity to the spin-liquid, the ED calculations on Model 1 (Fig. 2(c)) show sharp dispersive modes appearing over the majority of the Brillouin zone that are consistent with the conventional magnons of LSWT (Fig. 2(b)). Indeed, for energies below the main spin-wave branch ($\omega = 1.3 - 2.3$ meV), intensity is localized around the M- and Y-points, corresponding to the pseudo-Goldstone modes of the zigzag order (Fig. 2 (d)). ED calculations show clear spin-wave “cones” emerging from such points and extending to higher energies. Large deviations from LSWT are observed only for the highest energy excitations, which appear near the 2D Γ -point for energies $\omega > 5$ meV. Here, the ED calculations display a broad continuum-like feature centred at $\omega \sim K_1$ that resembles the response expected for the $K_1 > 0$ Kitaev spin-liquid, as shown in Fig. 2(e). However, comparison with the experimental $\mathcal{I}(\Gamma, \omega)$ shows poor agreement; while the experimental intensity extends from 2–15 meV, the ED results for Model 1 predict intensity only at high energies > 5 meV. Indeed, the evolution of the Γ -point intensity with $|J_1/K_1|$ is shown in Fig. 2(f). On approaching the $K_1 > 0$ spin-liquid by decreasing $|J_1/K_1|$, excitations at the Γ -point shift to higher energy, such that none of the parameters in the vicinity of the spin-liquid reproduce the experimental ω -dependence of $\mathcal{I}(\Gamma, \omega)$. Similar conclusions can also be drawn from recent DMRG studies of the nnHK model [32]. We therefore conclude that the broad features observed experimentally in $\mathcal{I}(\Gamma, \omega)$ at relatively low energies [12] are incompatible with pure Heisenberg and Kitaev interactions.

In order to treat the effect of interactions beyond the nnHK model, we consider now the *ab-initio*-guided Model 2. In contrast to Model 1, ED calculations on Model 2 (Fig. 3(c)), show large deviations from standard LSWT (Fig. 3(b)) over a wide range of \mathbf{k} and ω . In particular, sharp single-magnon-like peaks appear only near the pseudo-Goldstone modes at the M- and Y-points, matching with experimental observations [23]. Elsewhere, broad continuum-like features are observed within the ED resolution. As demonstrated in Fig. 3(d), we find significant intensity at low energies ($\omega < 2.3$ meV), at both the Γ - and (M,Y)-points, in agreement with the experiment. For the intermediate energy region ($\omega = 5.5 - 8.5$ meV), $\mathcal{I}(\mathbf{k})$ clearly resembles the six-fold star shape observed experimentally, while at higher energies ($\omega > 10.5$ meV) intensity is mainly located at the Γ -point, consistent with the experiment. Moreover, the ED results for the Γ -point intensity $\mathcal{I}(\Gamma, \omega)$ show a broad range of excitations peaked around 4 and 6 meV, and extending up to ~ 15 meV (Fig. 3(e)), in excellent agreement with the experiment [12]. Therefore, ED calcula-

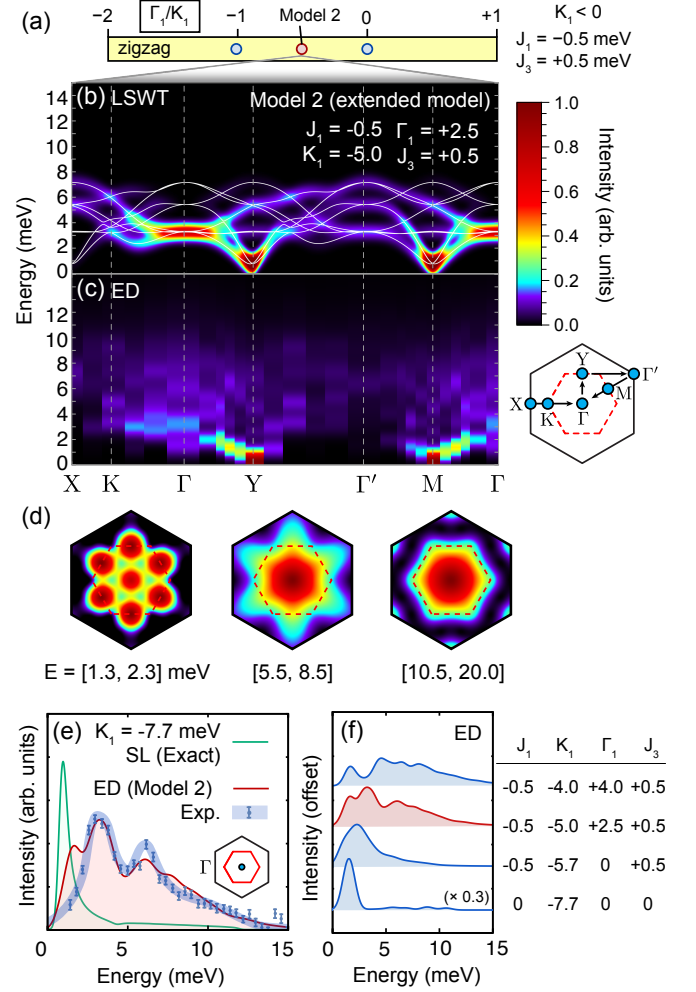


FIG. 3. Computed neutron scattering intensity $\mathcal{I}(\mathbf{k}, \omega)$ within the extended $(J_1, K_1, \Gamma_1, J_3)$ model. (a) Phase diagram for $K_1 < 0, J_3 = -J_1 = 0.5$ meV as a function of $|\Gamma_1/K_1|$, showing positions of interactions shown in (f). (b-d) Detailed results for Model 2: (b) $\mathcal{I}(\mathbf{k}, \omega)$ computed via LSWT; results are averaged over the three zigzag ordering wavevectors, parallel to the X, Y, and Z bonds. (c) ED results, combining data from several 20- and 24-site periodic clusters (see Methods). (d) ED \mathbf{k} -dependence of $\mathcal{I}(\mathbf{k}, \omega)$ integrated over the indicated energies, as obtained from a single 24-site cluster respecting all symmetries of Eq. (1) (see Methods). (e) Comparison of Γ -point intensities for the $K_1 < 0$ spin-liquid (exact results [30, 31]), Model 2 (ED), and the experimental data for α -RuCl₃ [12]. (f) Evolution of the ED Γ -point intensity with decreasing $|\Gamma_1/K_1|$, showing significant broadening at finite Γ_1 . For all spectra, a Gaussian broadening of 0.5 meV has been applied.

tions on Model 2 reproduce all of the main experimental spectral features, validating the range of interactions indicated by *ab-initio* calculations. Interestingly, the spectral features at the Γ -point become dramatically sharper on approaching the $K_1 < 0$ spin liquid, as shown in the evolution of $\mathcal{I}(\Gamma, \omega)$ with the ratio $|\Gamma_1/K_1|$ (Fig. 3(f)).

This result reveals that the broad continuum is not directly associated with proximity to the Kitaev state.

To gain further insight into the contrasting results of Models 1 and 2, it is useful to consider the possible magnon decay channels in the zigzag ordered phase. At the level of LSWT, the spin-wave Hamiltonian takes the quadratic form $\mathcal{H}_2 = \sum_{\mathbf{k},m} \epsilon_{\mathbf{k},m} a_{\mathbf{k},m}^\dagger a_{\mathbf{k},m}$ in terms of magnon creation (annihilation) operators a^\dagger (a), where $\epsilon_{\mathbf{k},m}$ denotes the dispersion for the m th magnon band. In this harmonic approximation, the magnons represent sharp, well-defined excitations. However, the total magnon number $N_{tot} = \sum_{\mathbf{k},m} a_{\mathbf{k},m}^\dagger a_{\mathbf{k},m}$ is typically not a conserved quantity, such that the stability of magnons is not guaranteed. Indeed, quantum fluctuations beyond LSWT appear through higher order anharmonic decay terms that may mix the sharp single-magnon modes with the multi-magnon continuum [33–35]. Similar considerations also apply to the breakdown of other collective modes, such as phonons in anharmonic crystals [36, 37]. From this perspective, a large decay rate is expected for any single magnon mode that is energetically degenerate with the multi-particle continuum, unless there are specific symmetries guaranteeing that the two do not couple. It is therefore useful to reconsider the prerequisites for magnon breakdown in the presence of the strongly anisotropic interactions of Eq. (1).

We first consider the stability of magnons in the nnHK model. For pure J_1 and K_1 interactions, the total spin projections $S_{tot}^\gamma = \sum_i S_i^\gamma$ are conserved along the cubic axes $\gamma = \{x, y, z\}$ modulo two. Since the ordered moment also lies along one of the cubic axes in the zigzag phase [19, 38] (see Fig. 4(b)), the possible magnon decay channels are somewhat restricted. In the local picture, the relevant quantum fluctuations are local singlet $S_i^x S_j^x | \uparrow\downarrow \rangle = | \downarrow\uparrow \rangle$ and triplet $S_i^x S_j^x | \uparrow\uparrow \rangle = | \downarrow\downarrow \rangle$ fluctuations shown in Fig. 4(a), with $\Delta S_{tot}^z = 0$ and 2, respectively. In the magnon picture, the Hamiltonian can only contain even order terms (i.e. $\mathcal{H} = \mathcal{H}_2 + \mathcal{H}_4 + \dots$), analogous to conventional Heisenberg antiferromagnets with collinear ordered spins [33, 35]. For example, the fourth order decay process \mathcal{H}_4 mixes the one-magnon states with the three-magnon continuum ($\Delta N_{tot} = \pm 2$)

$$\mathcal{H}_4 = \sum_{1-4} V_{123}^4 a_1^\dagger a_2^\dagger a_3^\dagger a_4 \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 - \mathbf{k}_4) + H.c. \quad (2)$$

Here, the bold index ($\mathbf{n} \equiv \mathbf{k}_n, m_n$) labels both momentum and magnon band. As noted above, the effect of such terms depends crucially on the availability of low-energy three-magnon states in which to decay.

The density of three-magnon states for Model 1 is shown in Fig. 4(c). At each \mathbf{k} -point, the lowest energy three-magnon state $a_{\mathbf{q}_1}^\dagger a_{\mathbf{q}_2}^\dagger a_{\mathbf{q}_3}^\dagger | 0 \rangle$, (with $\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3 = \mathbf{k}$) is obtained by placing two particles in the pseudo-Goldstone modes at opposite M-points ($\mathbf{q}_1 + \mathbf{q}_2 = 0$),

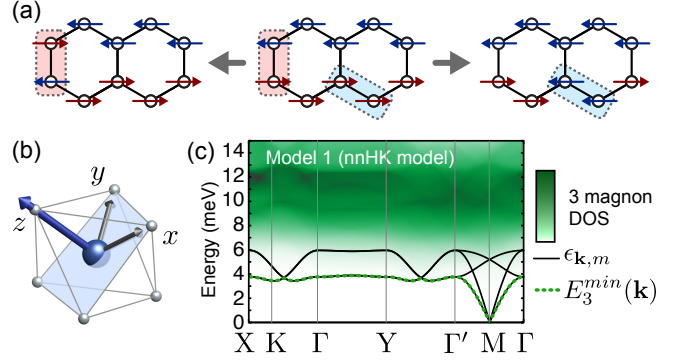


FIG. 4. Magnon decay channels for the nnHK model. (a) Local picture of quantum fluctuations away from zigzag order. (b) Ordered moment direction for zigzag ordering wavevector $\mathbf{Q} = \mathbf{Y}$, parallel to the Z-bond. (c) LSWT dispersions $\epsilon_{\mathbf{k},m}$, and 3-magnon density of states (DOS) for Model 1 with $\mathbf{Q} = \mathbf{Y}$. The dashed line indicates the bottom of the three-magnon continuum (E_3^{min}), which is coincident with the lowest magnon band.

and the third particle at $\mathbf{q}_3 = \mathbf{k}$, with total energy $E_3^{min}(\mathbf{k}) = \epsilon_{\mathbf{k},1} + 2\epsilon_{\mathbf{M},1}$. This implies $E_3^{min}(\mathbf{k}) \geq \epsilon_{\mathbf{k},1}$; the three-magnon states lie above the lowest one-magnon band at every \mathbf{k} -point. As a result, every magnon in the lowest band remains kinetically stable, due to the absence of low-energy three-particle states in which to decay. Precisely this condition ensures the stability of low-energy magnons in conventional isotropic antiferromagnets, and explains the sharp magnon-like peaks observed in Fig. 2(c) for Model 1. Strong spectral broadening in the nnHK model can occur only for high-lying excitations with $\epsilon_{\mathbf{k},m} > \epsilon_{\mathbf{k},1} = E_3^{min}$, where the density of three-magnon states is finite, such as at the 2D Γ -point. On approaching the spin-liquid (at $J_1 = 0$), this condition is relaxed due to the vanishing dispersion of the lowest magnon band (i.e. $\epsilon_{\mathbf{k},1} \rightarrow 0$), which corresponds to a vanishing energy cost for singlet fluctuations. Elsewhere, the majority of magnons are expected to remain stable.

In Model 2, the character of the quantum fluctuations is notably different (Fig. 5). The finite Γ_1 interaction reduces the local symmetry and leads to rotation of the ordered moments away from the cubic axes [19, 38] (Fig. 5(b)). In the local picture, this allows additional single-spin fluctuations $S_i^x S_j^z | \uparrow\uparrow \rangle = | \downarrow\uparrow \rangle$ (Fig. 5(a)), which correspond to odd-order anharmonic terms $\mathcal{H}_3, \mathcal{H}_5, \dots$ in the magnon Hamiltonian, where [34, 35]:

$$\mathcal{H}_3 = \sum_{1-3} \Lambda_{12}^3 a_1^\dagger a_2^\dagger a_3 \delta(\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3) + H.c. \quad (3)$$

At lowest order, such terms mix the single-magnon states with the two-magnon continuum ($\Delta N_{tot} = \pm 1$), shown in Fig. 5(c). For $\mathbf{Q} = \mathbf{Y}$, at each \mathbf{k} -point the lowest energy two-magnon state $a_{\mathbf{q}_1}^\dagger a_{\mathbf{q}_2}^\dagger | 0 \rangle$ is obtained by plac-

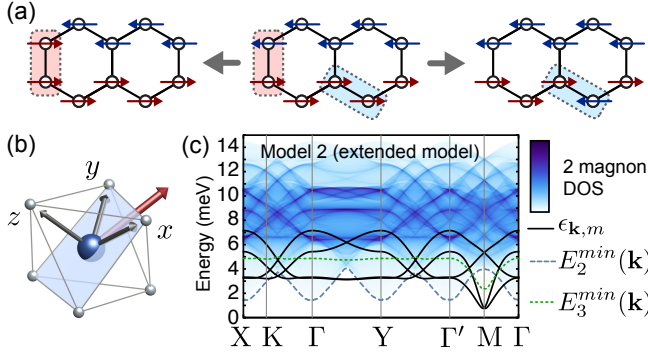


FIG. 5. Magnon decay channels for the extended (J_1, K_1, Γ_1, J_3) model. (a) Local picture of additional quantum fluctuations away from zigzag order induced by Γ_1 interactions. (b) Ordered moment direction for Model 2 with zigzag ordering wavevector $\mathbf{Q} = \mathbf{Y}$, parallel to the Z-bond. (c) LSWT dispersions $\epsilon_{\mathbf{k},m}$, and 2-magnon density of states (DOS) for Model 2 with $\mathbf{Q} = \mathbf{Y}$. Dashed lines indicate the bottom of the two- and three-magnon continuum ($E_2^{\min}(\mathbf{k})$ and $E_3^{\min}(\mathbf{k})$, respectively).

ing one particle in the pseudo-Goldstone mode at an M-point, and the second particle at $\mathbf{q}_2 = \mathbf{k} - \mathbf{M}$, with total energy $E_2^{\min}(\mathbf{k}) = \epsilon_{\mathbf{k}-\mathbf{M}} + \epsilon_{\mathbf{M}} \neq E_3^{\min}$. It should be emphasized that this condition differs from that of a conventional Heisenberg antiferromagnet (for which $E_2^{\min} = E_3^{\min}$) [35]. The difference is directly related to the strong anisotropic K_1 and Γ_1 interactions, which shift the pseudo-Goldstone modes to the M-points rather than the Γ -point or ordering wavevector \mathbf{Q} , as in conventional antiferromagnets [39]. This shift leads to an offset of the even and odd magnon states in \mathbf{k} -space. As a result, $E_2^{\min}(\mathbf{k}) < \epsilon_{\mathbf{k},1}$ over a wide region of the Brillouin zone; there are many two-magnon states with equal or lower energy than the one-magnon states. Provided there is a finite Γ_1 , the spontaneous decay of single magnons into the two-particle continuum is therefore allowed even for the lowest magnon band. For Model 2, the pseudo-Goldstone magnons near the M-points remain coherent due to the absence of low-energy two particle states in which to decay (Fig. 5(c)). This explains the experimental observation of sharp magnon-like modes near the M-points [23]. In contrast, the magnon bands in the remainder of the Brillouin zone directly overlap with the two-particle continuum. It is therefore natural to anticipate a large decay rate even at low energies.

The general requirements for strong two-magnon decays are less restrictive than proximity to a spin-liquid state. Indeed, a large decay rate is ensured by i) large anisotropic interactions, ii) deviation of the ordered moments away from the high-symmetry axes, and iii) the shifting of low-energy magnons away from the Γ -point. Experimentally, these conditions are likely also satisfied by the zigzag ordered Na_2IrO_3 [9], and spiral magnets α -, β -, and γ - Li_2IrO_3 [40–42]. This picture is consistent with

recent indications that the magnetically disordered phase observed at high pressure in β - Li_2IrO_3 [43] is driven primarily by large Γ_1 interactions [44].

Discussion – There are two general scenarios that can explain the observed continuum excitations in α - RuCl_3 and the iridates A_2IrO_3 . In the first scenario, the emergent excitations are weakly interacting, but possess quantum numbers (e.g. $\Delta S_{\text{tot}} = \pm 1/2$) inconsistent with the experimental selection rules ($\Delta S_{\text{tot}} = 0, \pm 1$). Such fractional excitations must be created in multiples experimentally, but can otherwise be treated as free particles, leading to a *coherent* multi-particle continuum in the observed spectra. This scenario describes the Kitaev spin-liquid, where the special symmetries of the Hamiltonian allow an exact description in terms of non-interacting Majorana spinons and localized fluxes [5]. However, away from the pure Kitaev point, the relevant symmetries are lifted both by the additional magnetic interactions, and the spontaneous symmetry breaking of the magnetic order, which tends to confine the spinons into gauge neutral objects such as magnons [45, 46]. Despite this latter tendency, we have argued that coherent magnons are unlikely to appear at large Γ_1 , due to the strong anharmonicity in the magnon Hamiltonian. While it remains possible that nearly free Majorana spinons persist into the zigzag ordered phase, a more general second scenario is that the observed continua represent *incoherent* excitations arising from the complex, low-symmetry interactions. Such strongly correlated excitations may not be describable in terms of free particles, either conventional or exotic. At present, it is not clear which of these scenarios applies to the iridates and α - RuCl_3 , although a key role must be played by both the Kitaev K_1 and off-diagonal couplings such as Γ_1 .

Conclusion – In summary, we have shown that all main features of the magnetic excitations in α - RuCl_3 [12, 13, 23] are consistent with strongly anisotropic interactions having signs and relative magnitudes in agreement with *ab-initio* predictions. The ferromagnetic Kitaev coupling ($K_1 < 0$) is supplemented by a significant off-diagonal term ($\Gamma_1 > 0$) that plays a crucial role in establishing both the zigzag order and the observed continua. In the presence of such interactions, the conventional magnon description breaks down even deep in the ordered phase, due to strong coupling of the one-magnon and two-magnon states. This effect is expected to persist over a large range of the phase diagram suggesting the observed continua in α - RuCl_3 and the iridates A_2IrO_3 represent a rich and general phenomenon extending beyond the Kitaev spin-liquid. For this class of strongly spin-orbital coupled magnets, the presence of complex and frustrated anisotropic interactions leads naturally to dominant anharmonic effects in the inelastic magnetic response. Fully describing the dynamics of these and similar materials therefore represents a formidable challenge that is likely to reveal novel aspects not found in

conventional isotropic magnets.

Methods – The neutron scattering intensity was computed via:

$$\mathcal{I}(\mathbf{k}, \omega) \propto f^2(\mathbf{k}) \int dt \sum_{\mu, \nu} (\delta_{\mu, \nu} - k_{\mu} k_{\nu} / k^2) \times \quad (4)$$

$$\times \sum_{i, j} \langle S_i^{\mu}(t) S_j^{\nu}(0) \rangle e^{-i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j) - i\omega t}$$

where $f(\mathbf{k})$ is the atomic form factor of Ru^{3+} from Ref. [47]. ED calculations were performed using the Lanczos algorithm [48], on several 20- and 24-site clusters with periodic boundary conditions. Such periodic clusters are detailed in the supplemental material. Excitations were computed using the continued fraction method [49]. Further details and additional results are presented in the supplemental material; these extensive calculations go beyond previous ED studies [15, 16, 19, 25, 28], which focussed mainly on the static properties, or a limited portion of the phase diagram. ED results shown for the high-symmetry Γ , M, Y, X, and Γ' points were averaged over all clusters. ED \mathbf{k} -dependence of $\mathcal{I}(\mathbf{k}, \omega)$, integrated over the energy windows $E = 1.3 - 2.3$, $5.5 - 8.5$, and $10.5 +$ meV (Fig. 2(d) and 3(d)), were obtained from a single 24-site cluster respecting all symmetries of the model. In all cases, the discrete ED and LSWT spectra were Gaussian broadened by 0.5 meV, consistent with the width of experimental features [12]. The intensities were also averaged over the same range of out-of-plane momentum as in the experiment [12].

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